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Claims

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- 1. A curable composition, comprising:
- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is essentially free of bromine atoms;
- (b) a flame retardant additive essentially free of phenolic groups and of epoxy groups, wherein said flame retardant is a condensation product of (i) a brominated phenol or a mixture of brominated phenols with (ii) a cyanuric halide;
- (c) a thermoplastic resin; and
- (d) a cyanate ester.

[c2]

2. The curable composition of claim 1, wherein said flame retardant additive has a bromine content greater than 20%.

[c3]

3. The curable composition of claim 1, wherein said flame retardant additive is 1,3,5-tris(2,4,6-tribromophenoxy)triazine.

[c4]

4. The curable composition of claim 1, wherein said flame retardant additive is 2,2'-[(1-methylethylidene)bis[(2,6-dibromo-4,1-phenylene)oxy]]bis[4,6-bis [(2,4,6-tribromophenyl)oxy]-1,3,5-triazine].

[c5]

5. The curable composition of claim 1, wherein said flame retardant additive is soluble in toluene at a concentration of greater than 15 g/100ml of toluene at a temperature of 50 $^{\circ}$ C.

[c6]

6. The curable composition of claim 1, wherein said epoxy resin is a glycidyl ether resin or a mixture of glycidyl ether resins containing, on average, greater than 2 epoxy groups per molecule.

[c7]

7. The curable composition of claim 1, wherein said epoxy resin is a mixture of:
(a1) an epoxy resin containing on average less than or equal to 2 glycidyl groups per molecule; and
(a2) an epoxy resin containing greater than 2 glycidyl groups per molecule.

[c8]

8. The curable composition of claim 1, wherein said thermoplastic resin has a

Tg greater than 120 ° C.

[c9]	9. The curable composition of claim 1, wherein said thermoplastic resin has a dissipation factor of less than 0.010 measured at 1 MHz at room temperature.
[c10]	10. The curable composition of claim 1, wherein said thermoplastic resin has been directly isolated from solution after polymerization.
[c11]	11. The curable composition of claim 1, wherein said thermoplastic resin is a poly(phenylene ether).
[c12]	12. The curable composition of claim 11, wherein said poly(phenylene ether) has a weight average molecular weight ranging from about 3,000 to 35,000 g/mol.
[c13]	13. The curable composition of claim 11, wherein said poly(phenylene ether) has a weight average molecular weight ranging from about 3,000 to 35,000 g/mol.
[c14]	14. The curable composition of claim 11, wherein said poly(phenylene ether) has been melt processed at a temperature ranging from about 200 ° to 350 ° C.
[c15]	15. The curable composition of claim 11, wherein said poly(phenylene ether) is hydroxy functional.
[c16]	16. The curable composition of claim 1, wherein said thermoplastic resin is one or more of a poly(phenylene ether) or a poly(styrene- co -maleic anhydride).
[c17]	17. The curable composition of claim 1, wherein said thermoplastic resin is a reaction product of a poly(phenylene ether) and a peroxide.
[c18]	18. The curable composition of claim 1, wherein said thermoplastic resin is a reaction product of a poly(phenylene ether), a peroxide, and a bisphenol.
[c19]	19. The curable composition of claim 1, wherein said thermoplastic resin is a polyimide.
[c20]	20. The curable composition of claim 1, wherein the curable composition further comprises one or more of an organic reinforcement, an inorganic

[c24]

reinforcement, or a filler.

- [c21] 21. The curable composition of claim 1, wherein the curable composition is essentially free of homopolymers of styrene.
- [c22] 22. The curable composition of claim 1, wherein the epoxy resin is a multifunctional glycidyl ether.
- [c23] 23. The curable composition of claim 22, wherein said multifunctional glycidyl ether is selected from the group consisting of epoxidized phenol–formaldehyde novolacs, epoxidized cresol–formaldehyde novolacs, epoxidized alkylphenol–formaldehyde novolacs, epoxidized 1,1,1-tris(4-hydroxyphenyl)ethane, epoxidized 1,1,2,2-tetra(4-hydroxyphenyl) ethane, epoxidized phenol–dicyclopentadiene novolacs, and epoxidized phenol–benzaldehyde novolacs.
 - 24. The curable composition of claim 1, wherein the cyanate ester is selected from the group consisting of 1,3- dicyanatobenzene, 1,4-dicyanatobenzene, 2tert-butyl-1,4-dicyanatobenzene, 2,4-dimethyl-1,3-dicyanatobenzene, 2,5-ditert-butyl-1,4-dicyanatobenzene, tetramethyl-1,4-dicyanatobenzene, 4chloro-1,3-dicyanatobenzene, 1,3,5-tricyanatobenzene, 2,2'dicyanatobiphenyl, 4,4'-dicyanatobiphenyl, 3,3',5,5'-tetramethyl-4,4'dicyanatobiphenyl, 1,3-dicyanatonaphthalene, 1,4-dicyanatonaphthalene, 1,5dicyanatonaphthalene, 1,6-dicyanatonaphthalene, 1,8-dicyanatonaphthalene, 2,6-dicyanatonaphthalene, 2,7-dicyanatonaphthalene, 1,3,6tricyanatonaphthalene, bis(4-cyanatophenyl)methane, bis(3-chloro-4cyanatophenyl)methane, bis(3,5-dimethyl-4-cyanatophenyl)methane, 1,1-bis (4-cyanatophenyl)ethane, 2,2-bis(4-cyanatophenyl)propane, 2,2-bis(3,3dibromo-4-cyanatophenyl)propane, 2,2-bis(4-cyanatophenyl)-1,1,1,3,3,3hexafluoropropane, bis(4-cyanatophenyl)ester, bis(4-cyanatophenoxy)benzene, bis(4-cyanatophenyl)ketone, bis(4-cyanatophenyl)thioether, bis(4cyanatophenyl)sulfone, tris(4-cyanatophenyl)phosphate, and tris(4cyanatophenyl)phosphate.
- [c25] 25. The curable composition of claim 1, wherein the cyanate ester has the formula $R \left(O C \equiv N\right)_n$

wherein R is an aromatic nucleus-containing residue which is selected from the group consisting of a residue derived from an aromatic hydrocarbon selected from the group consisting of benzene, biphenyl and naphthalene, a residue derived from a compound in which at least two benzene rings are bonded to each other by a bridging member selected from the group consisting of

wherein R $_{\rm a}$ and R $_{\rm b}$ are the same or different and each represents a hydrogen atom or an alkyl group containing 1 to 4 carbon atoms,

$$-0-$$
 , $-CH_2OCH_2-$, $-S-$, $-C-$, $-S-$, $-S-$, $-O-P-O-$ 0

and

and a residue resulting from the removal of a phenolic hydroxyl group from a novolac-type or resol-type phenolic resin skeleton; said aromatic nucleus is optionally substituted by a substituent selected from the group consisting of alkyl groups containing 1 to 4 carbon atoms, alkoxy groups containing 1 to 4 carbon atoms, chlorine and bromine; n is an integer of 2 to 5; and the cyanate group is always directly bonded to the aromatic nucleus.

[c26]

26. The curable composition of claim 1, wherein the cyanate ester is a prepolymer of of the cyanates esters of Claim 25, having a number average molecular weight of 400 to 6,000, and are formed by trimerizing the cyanate group of the cyanate esters.

[c27]

27. The curable composition of claim 1, wherein the cyanate ester is a cyanate-group-containing phenol resin comprising a mixture of polymers represented by the formula

wherein n is 0 or an integer of 1 or more; and R's may be the same or different, and each R is a hydrogen atom or a methyl group, and containing 50% by weight or more in total of polymers having formula in which n is an integer of 1 to 3, the number average molecular weight of said phenol resin being 350 to 700 g/mole.

[c28]

28. The curable composition of claim 1, wherein the cyanate ester is a cyanic acid ester of an aromatic polycarbonate obtained by reacting an aromatic polycarbonate having one or two terminal hydroxyl groups with a cyanogen halide, wherein the aromatic polycarbonate is prepared from an aromatic dihydroxy compound represented by one of the following formulas:

where n is an integer of 1-4, inclusive, or a mono-, di-, tri- or tetra- halogeno-

nuclear substituted derivative of the aromatic dihydroxy compound represented by one of the above formulas.

[c29] 29. The curable composition of claim 1, wherein the cyanate ester is a cyanatophenyl-terminated polyarylene ether of the formula

where R is a divalent radical having 3 to 15 aromatic nuclei linearly linked together with ethereal oxygen atoms, said nuclei comprising nuclei selected from the group consisting of

[c30]

30. The curable composition of claim 1, wherein the cyanate ester has the structure

$$H_3C$$
 CH_3
 OCN
 H_4C
 CH_4

wherein X is methylene, isopropylidene, oxygen or divalent sulfur.

[c31]

31. The curable composition of claim 1, wherein the cyanate ester is a blend of a tricyanate ester and a dicyanate ester, wherein the tricyanate ester has the structural formula:

and the dicyanate ester has the structural formula:

wherein each R is H or methyl and is the same or different and wherein X is methylene, alkylidene having 2 to 4 carbon atoms, divalent oxygen, or divalent sulfur.

[c32]

32. The curable composition of claim 1, wherein the cyanate ester is a polyaromatic cyanate having the formula

wherein: E is an aromatic radical;

B is a C $_{7-20}$ polycyclic aliphatic radical;

D is independently in each occurrence any nonactive hydrogen-containing substituent;

q, r and s are independently in each occurrence the integers 0, 1, 2, or 3; with

the proviso that the sum of q, r and s is greater than or equal to 2; t is independently in each occurrence an integer of between about 0 and 4 inclusive; and

x is a number between about 0 and 5 inclusive.

[c33] 33. The curable composition of claim 1, wherein the cyanate ester is a fluorocarbon monocyanate having the structure $F_3C(CFX)_aA(CFX)_bCH_2OCN$

where A is

$$-(o-c_{FX}-c_{F_2}-o)$$

X is fluorine or perfluoroalkyl having 1 to 10 carbon atoms, a is 1 to 10, h is 1, and c is 1 to 100.

[c34] 34. The curable composition of claim 1, wherein the cyanate ester is a fluorocarbon dicyanate having the structure NCOCH₂(CFX)_aB(CFX)_bCH₂OCN

where B is (I) a carbon-to-carbon bond, in which case a is an integer of 1 to 30 and b is zero, or (II) B is [(CFX) $_{\rm d}$ O(CFX) $_{\rm e}$ $_{\rm f}$, in which case a and b are zero, d and e are integers of 1 to 30, and f is an integer of 1 to 20, or (III) B is $_{\rm CCF_2-CFX}$ $_{\rm g}$ O(CFX) $_{\rm h}$ O(CFX-CF₂O) $_{\rm f}$

in which case a and b are 1, h is an integer of 1 to 10, and g and i are integers of 1 to 100, or (IV) B is $[(CF_2CH_2)_i(CFZ-CFX)_k]_m,$

in which case a and b are integers of 1 to 10, j and k are integers whose ratio j/k is 1/1 to 10/1, m is an integer of 1 to 100, and (CF $_2$ CH $_2$) and (CF $_2$ -CFX) are randomly distributed units; and where X in all instances where it appears is fluorine or perfluoroalkyl of 1 to 10 carbon atoms.

[c35] 35. The curable composition of claim 1, wherein the cyanate ester has the formula

$$(NCO)_d$$
 A
 $(R)_b$
 $(R)_c$

in which R represents hydrogen, halogen, linear or branched C $_1$ -C $_9$ -alkyl or phenyl, two adjacent radicals R on the same nucleus together forming a

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carbocyclic 5-membered or 6-membered ring or together and in conjunction with a hetero atom (O, S, N) forming a 5-membered or 6-membered heterocyclic ring, alkoxy radicals with 1 to 4 carbon atoms, alkoxy carbonyl radicals with 1 to 4 carbon atoms in the alkyl group;

R' has the same meaning as R or represents the group

$$-A - (OCN)_d$$

$$(R)_b$$

where A is direct bond, a C $_1$ -C $_9$ -alkylene group optionally substituted by C $_1$ -C $_4$ -alkyl or phenyl, a cycloaliphatic or aromatic 5-membered or 6-membered ring, or a cycloaliphatic or aromatic 5-membered or 6-membered ring; a is a number from 0 to 5 where e = 1 and a number from 2 to 5 where e = 0; b = 5 - a where e = 1 and 6 - (a + d) where e = 0; c = 5 - d; d is a number from 0 to 5; e is the number 0, 1, 2 or 3; with the proviso that the sum of a and d (a + d) always gives a number from 2 to 5.

[c36]

36.A curable composition, comprising:

(a)an epoxy resin and curing agent therefor, wherein said epoxy resin is a glycidyl ether resin or mixture of glycidyl ether resins containing, on average, greater than 2 epoxy groups per molecule;

(b)1,3,5-tris(2,4,6-tribromophenoxy)triazine and/or 2,2'-[(1-methylethylidene) bis[(2,6-dibromo-4,1-phenylene)oxy]]bis[4,6-bis[(2,4,6-tribromophenyl)oxy]-1,3,5-triazine];

(c)a poly(phenylene ether) resin; and (d)a cyanate ester.

[c37]

37.A curable composition, comprising:

(a)an epoxidized cresol-formaldehyde novolac resin;

(b)1,3,5-tris(2,4,6-tribromophenoxy)triazine;

(c)a poly(phenylene ether) resin having a number average molecular weight ranging from about 1,000 to 15,000 g/mol; and (d)a cyanate ester.

[c38]

38.A cured composition comprising a cured residue of a curable composition comprising:

- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is essentially free of bromine atoms;
- (b)a flame retardant additive essentially free of phenolic groups and of epoxy groups, wherein said flame retardant is a condensation product of (i) a brominated phenol or a mixture of brominated phenols with (ii) a cyanuric halide;
- (c)a thermoplastic resin; and
- (d) a cyanate ester.